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TITLE : LIGHT RESISTANT POLYCARBONATE-BASED RESIN COMPOSITION

ABSTRACT : PROBLEM TO BE SOLVED: To obtain the subject composition not yellowing by exposure to light over a long period of time, having a stable color tone and suitable for an trimming part of an automobile, etc., by simultaneously using a specific yellow dyestuff with a light-stabilizer.

SOLUTION: This light resistant polycarbonate-based resin composition is obtained by blending 100 pts.wt. polycarbonate-based resin consisting of (A) 1-100wt.% polycarbonate resin and (B) 0-99wt.% thermoplastic resin capable of mixing and mulling with the component (A), with (C) 0.05-2 pt.wt. light- stabilizer and (D) 0.0005-0.2 pt.wt. yellow dyestuff having a property of changing to white color on exposure to light. Further, it is preferable to use a polybutyleneterephthalate, a polyethyleneterephthalate, an acrylonitrile-EPDM- styrene copolymer or an acrylonitrile-butadiene-styrene copolymer as the component (B), a compound having a benzotriazole, a benzophenone or a piperidine skeleton in molecule thereof as the component (C) and solvent yellow 157 of a color index number as the component (D).

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JAPANESE / ENGLISH TRANSLATION OF

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Title: Lightfast Polycarbonate Resin Composition

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(54) [Title of the Invention]

Lightfast Polycarbonate Resin Composition

(57) [Summary]

[Constitution] A lightfast polycarbonate resin composition obtained as a result of compounding 0.05 to 2 weight parts of a photostabilizer (C) and 0.005* to 0.2 weight parts of a yellow dyestuff (D) that possesses the property of being able to turn white when exposed to light, with 100 weight parts of a polycarbonate resin comprising 1 to 100 % by weight of a polycarbonate

* [Translator's note: This is presumably an error for "0.0005", which appears in the Claims and Means sections.]

resin (A) and 0 to 99 % by weight of a thermoplastic resin able to be kneaded with the polycarbonate resin.

[Effect] The invention exhibits minimal variation in color when exposed to light, retains a stable color tone over prolonged periods, and may be suitably used in outdoor applications or in other environments where exposure to light occurs.

[Claims]

[Claim 1] A lightfast polycarbonate resin composition obtained as a result of compounding 0.05 to 2 weight parts of a photostabilizer (C) and 0.0005 to 0.2 weight parts of a yellow dyestuff (D) that possess the property of being able to turn white when exposed to light, with 100 weight parts of a polycarbonate resin comprising 1 to 100 % by weight of a polycarbonate resin (A) and 0 to 99 % by weight of a thermoplastic resin able to be kneaded with the polycarbonate resin.

[Claim 2] The lightfast polycarbonate resin composition according to claim 1, characterized in that the thermoplastic resin (B) is polybutylene terephthalate, polyethylene terephthalate, acrylonitrile-EPDM-styrene (AES) copolymer, or acrylonitrile-butadiene-styrene copolymer (ABS).

[Claim 3] The lightfast polycarbonate resin composition according to claim 1 or 2, characterized in that the photostabilizer (C) is a compound whose molecules have a benzotriazole, benzo-phenone, or piperidine backbone.

[Claim 4] The lightfast polycarbonate resin composition according to claim 1, 2 or 3, characterized in that the yellow dyestuff (D) has color index numbers Solvent Yellow 157, Solvent Green 5, Disperse Yellow 201, Disperse Orange 47, Pigment Yellow 180, or Pigment Orange 43.

[Detailed Description of the Invention]

[0001]

[Field of Industrial Application] The present invention relates to a lightfast polycarbonate resin composition. More specifically, the invention provides a polycarbonate resin composition that is impervious to color variation when exposed to light, and retains a stable color tone over prolonged periods.

[0002]

[Prior Art] Polycarbonate resins, and polycarbonate alloys comprising such polycarbonate resins and thermoplastic resins that can be kneaded with these polycarbonate resins, are problematic in that their color changes under exposure to light and assumes a yellowish hue (i.e., yellowing occurs). Consequently, when articles formed such resins are exposed for prolonged periods outdoors or in other environments, problems are encountered when light-induced yellowing phenomena occur, and the color tone of the molded articles changes. Attempts employed in the past to resolve these problems have involved compounding a UV absorber or other photostabilizer with the polycarbonate resin or polycarbonate alloy.

[0003]

[Problems to Be Resolved by the Invention] Nevertheless, it has been impossible to prevent the molded articles from yellowing when exposed to light using this method alone. The present invention resolves these problems.

[0004]

[Means for Solving the Aforementioned Problems] The present inventors conducted assiduous investigations into resolving the aforementioned problems, and accordingly perfected the present invention based on the discovery that the use of a specific yellow dyestuff in concert with a photostabilizer stably preserves the color tone and prevents yellowing, even under exposure to light for prolonged periods.

[0005] In other words, the present invention provides a lightfast polycarbonate resin composition obtained as a result of compounding 0.05 to 2 weight parts of a photostabilizer (C) and 0.0005 to 0.2 weight parts of a yellow dyestuff (D) that possess the property of being able to turn white when exposed to light, with 100 weight parts of a polycarbonate resin comprising 1 to 100 % by weight of a polycarbonate resin (A) and 0 to 99 % by weight of a thermoplastic resin able to be kneaded with the polycarbonate resin.

[0006] The present invention shall be described in further detail hereunder. The polycarbonate resin (A) used in the present invention is a polymer obtained by means of phosgenation, in which a variety of dihydroxydiaryl compounds are reacted with a phosgene; or transesterification, in which a dihydroxydiaryl compound is reacted with diphenyl carbonate or another carbonate,

with a typical example being a polycarbonate resin manufactured from 2,2-*bis*(4-hydroxyphenyl)propane (commonly known as "bisphenol A").

[0007] Examples of other dihydroxydiaryl compounds besides bisphenol A include *bis*(4-hydroxyphenyl)methane, 1,1-*bis*(4-hydroxyphenyl)ethane, 2,2-*bis*(4-hydroxyphenyl)butane, 2,2-*bis*(4-hydroxyphenyl)octane, *bis*(4-hydroxyphenyl)phenylmethane, 2,2-*bis*(4-hydroxyphenyl-3-methylphenyl)propane, 1,1-*bis*(4-hydroxy-3-*tert*-butylphenyl)propane, 2,2-*bis*(4-hydroxy-3-bromophenyl)propane, 2,2-*bis*(4-hydroxy-3,5-dibromophenyl)propane, 2,2-*bis*(4-hydroxy-3,5-dichlorophenyl)propane, and other *bis*(hydroxyaryl)alkanes; 1,1-*bis*(4-hydroxyphenyl)cyclopentane, 1,1-*bis*(4-hydroxyphenyl)cyclohexane, and other *bis*(hydroxyaryl)cycloalkanes; 4,4'-dihydroxydiphenyl ether, 4,4'-dihydroxy-3,3'-dihydroxydiaryl ether, and other 3,3'-dimethyldiphenyl ethers; 4,4'-dihydroxydiphenyl sulfide and other dihydroxydiaryl sulfides; 4,4'-dihydroxydiphenyl sulfoxide, 4,4'-dihydroxy-3,3'-dimethyldiphenyl sulfoxide, and other dihydroxydiaryl sulfoxides; and 4,4'-dihydroxydiphenyl sulfone, 4,4'-dihydroxy-3,3'-dimethyldiphenyl sulfone, and other dihydroxy diaryl sulfones.

[0008] These compounds may be used singly, in combinations of two or more, or blended with piperazine, dipiperidyl hydroquinone, resorcinol, 4,4'-dihydroxydiphenyl, and the like.

[0009] The dihydroxyaryl [sic] compounds may be blended with the following phenolic compounds that have a valence of three or higher. Examples of phenols having a valence of three or higher include fluoroglucine, 4,6-dimethyl-2,4,6-tri(4-hydroxyphenyl)-heptene, 2,4,6-dimethyl-2,4,6-tri(4-hydroxyphenyl)-heptane, 1,3,5-tri(4-hydroxyphenyl)-benzole, 1,1,1-tri(4-hydroxyphenyl)-ethane, and 2,2-*bis*-[4,4-(4,4'-dihydroxydiphenyl)-cyclohexyl]-propane.

[0010] There are no particular limitations as to the viscosity-average molecular weight of the polycarbonate resin (A), but due to considerations of moldability/workability and strength, common levels are 10000 to 100000, and preferably 15000 to 35000. A molecular weight [sic], catalyst, or the like may be used if necessary when manufacturing the polycarbonate resin.

[0011] Examples of the thermoplastic resin (B) used in the present invention include polybutylene terephthalate, polyethylene terephthalate, and other aromatic polyesters, as well as polystyrene (PS), high-impact polystyrene (HIPS), styrene-acrylonitrile copolymer (SAN), acrylonitrile-butadiene-styrene copolymer (ABS), acrylonitrile-EPDM-styrene copolymer (AES), and other styrene-based copolymers. These resins may be used singly or in combinations of two

or more. At least one thermoplastic resin selected from among polybutylene terephthalate, polyethylene terephthalate, and AES resin is preferably used. When blended with polycarbonate resin (A), the thermoplastic resin (B) may account for 0 to 99 % by weight of the resulting mixture, with the content thereof preferably being 0 to 60 % by weight, and more preferably 0 to 45 % by weight.

[0012] Examples of the photostabilizer (C) used in the present invention include benzotriazole-, benzophenone-, and piperidine-based photostabilizers. These photostabilizers may be used singly or in combinations of two or more.

[0013] 0.05 to 2 weight parts of the photostabilizer (C) is used per 100 weight parts of either the polycarbonate resin (A) or a mixture comprising this polycarbonate resin and the thermoplastic resin (B). It is not desirable for less than 0.05 weight parts to be used, since poor lightfastness will result, or for more than two weight parts to be used, since the effect will reach a level of saturation. A more suitable range is 0.3 to 1 weight parts.

[0014] The yellow dyestuff (D) used in the present invention has the capacity to whiten when exposed to light, with examples of yellow dyestuffs exhibiting such a property including Solvent Yellow 157, Solvent Green 5, Disperse Yellow 201, Disperse Orange 47, Pigment Yellow 180, and Pigment Orange 43. These dyestuffs may be used singly or in combinations of two or more.

[0015] The amount of the yellow dyestuff (C) [sic] used is 0.0005 to 0.2 weight parts per 100 weight parts of either the polycarbonate resin (A) or a mixture comprising the polycarbonate resin and the thermoplastic resin (B). It is not desirable for less than 0.0005 weight parts to be used, since poor lightfastness will result, or for more than 0.2 weight parts to be used, since the decolorizing effect will become excessive, and inevitably lead to variations in colortone. A range of 0.001 to 0.1 weight parts is preferable. A range of 0.001 to 0.003 weight parts is more preferable.

[0016] There are no particular limitations as to the method employed to blend the polycarbonate resin (A), the thermoplastic resin (B) that is able to be kneaded with the polycarbonate resin, the photostabilizer (C), and the yellow dyestuff (D) that exhibits the capacity to whiten when exposed to light; for example, blending may be performed using a tumbler, ribbon blender, high-speed mixer, or other desired mixer, and melt-kneading may be performed using an extruder or the like.

[0017] When necessary, a release agent, filler, Impact modifier, antistatic agent, antioxidant, flame-retardant, dyestuff, pigment, or other additive may be introduced during mixing.

[0018] The lightfast polycarbonate resin composition of the present invention may be suitably used in components used for automobile exteriors (e.g., hubcaps, door handles, bumpers, and door mirrors), or other applications where the composition will be used outdoors or otherwise be exposed to light.

[0019]

[Working Examples] The present invention is described in detail below with reference to working examples; however, the present invention shall not be construed to be limited to these examples. Unless expressly indicated otherwise, references to "parts" and "%" in the examples respectively designate parts by weight and percent by weight.

[0020] Testing method

The resulting plate samples were subjected to irradiation for 200 hrs and 400 hrs at a black panel temperature of 63°C and under falling rain using a weathermeter ("WEL-SUN-HCH-B" Sunshine Super Long-life Weathermeter; manufactured by Suga Test Instruments Co., Ltd.).

[0021] Evaluation method

The change in hue before and after irradiation was obtained with the following formulae as a result of determining the respective stimulus values X, Y, and Z for red, green, and blue under a D65 light source (correlated color temperature: approximately 6504 K) with the aid of a spectrophotometer ("CMS-35SP"; manufactured by Murakami Color Research Laboratory), and determining the degree of change in yellowness; i.e., the yellowness index (ΔYI), with regard to the pre-irradiated plate samples.

$$\Delta YI = YI_0 - YI_1$$

$$YI = 100(1.28X - 1.06Z)/Y$$

YI_0 : plate sample before irradiation

YI_1 : plate sample after irradiation

The color difference (ΔE^*) is a measure of a color shift relative to the plate sample before irradiation, and is calculated using the following formula after the tristimulus values X, Y, and Z (determined using the spectrophotometer as described in the foregoing) have been converted to

L* (lightness), a* (chromaticity from reddish to greenish), and b* (chromaticity from yellowish to bluish).

$$\Delta E^* = \sqrt{((L_0^* - L_1^*)^2 + (a_0^* - a_1^*)^2 + (b_0^* - b_1^*)^2)}$$

L₀*, a₀*, b₀*: plate sample before irradiation

L₁*, a₁*, b₁*: plate sample after irradiation

In other words, the closer ΔYI (yellowness index) and ΔE* (color difference) get to zero, the closer the hue will be to the pre-irradiation level, and the less that the color will have changed under irradiation with light.

[0022] Working Example 1

A tumbler was used to dry-blend the components shown in Table 1 with 1.05 parts of titanium oxide white (RCL-128; manufactured by SCM) and 0.01 parts of carbon black (MA-600B; manufactured by Mitsubishi Chemical Corporation) as colorants. A 40-mm single-screw extruder was used to melt-blend the resulting mixture at a preset temperature of 250°C, yielding pellets of composition 1. The resulting pellets 1 were injection-molded at a preset temperature of 250°C by means of using a four-ounce injection molding machine in order to produce a 3.2 × 55 × 45 mm plate sample 1. The lightfastness of the resulting plate sample 1 was evaluated according to the aforementioned test and evaluation methods. The results are displayed in Table 1. Composition 2 and plate sample 2 were obtained as comparative examples following the same procedure. The color tone of composition 2 was adjusted to the same tone as composition 1 by means of using titanium oxide white and carbon black. The results are displayed in Table 1.

[0023]

[Table 1]

	Working Example	Comparative Example
Plate sample	1	2
Polycarbonate resin (A) *1	100 parts	100 parts
Photostabilizer (C) *2	1.5 parts	—
*3	0.2 parts	—
Yellow dyestuff (D) *4	0.005 parts	0.005 parts
Solvent Yellow 157		
ΔYI 200 hrs	2.2	16.8
400 hrs	2.4	21.0
ΔE* 200 hrs	1.2	7.9
400 hrs	1.6	9.8

*1: Polycarbonate resin (200-13); manufactured by Sumitomo Dow, Ltd.

*2: Benzotriazole-based photostabilizer (Chemisorb 79; manufactured by Chemipro Kasei Kaisha, Ltd.)

*3: Piperidine-based photostabilizer (Chimasorb 944FD; manufactured by Ciba-Geigy Corporation)

*4: Sumiplast Lemon Yellow HGN (manufactured by Sumitomo Chemical Co., Ltd.)

[0024] Working Example 2

The same procedure as described in Working Example 1 was employed in order to obtain compositions 3 through 6 and plate samples 3 through 6, with the exception that the components shown in Table 2 were used, in the composition ratios showed therein. The results of each lightfastness evaluation are displayed in Table 2.

[0025]

[Table 2]

Plate sample	Working Example		Comparative Example	
	3	4	5	6
Polycarbonate resin (A) *1	50 parts	65 parts	50 parts	65 parts
AES resin (B) *5	50 parts	—	50 parts	—
ABS resin (B) *6	—	35 parts	—	35 parts
Photostabilizer (C) *2	0.5 parts	0.5 parts	—	—
*3	0.5 parts	0.5 parts	—	—
Yellow dyestuff (D)				
Solvent Yellow 157 *4	0.004 parts	0.005 parts	0.004 parts	0.005 parts
ΔYI 200 hrs	2.0	2.4	9.6	10.2
400 hrs	2.6	2.9	13.6	15.3
ΔE^* 200 hrs	1.1	1.3	5.1	5.8
400 hrs	1.3	1.6	7.0	8.5

*5: AES resin (UB-500P; manufactured by Sumika ABS Latex Company)

*6: ABS resin (K-2540A; manufactured by Sumika ABS Latex Company)

[0026] Working Example 3

The same procedure as described in Working Example 1 was employed in full in order to obtain compositions 7 and 8 and plate samples 7 and 8, with the exception that 0.5 parts of titanium oxide white and 1.4 parts titanium of oxide yellow (TY-70; manufactured by Ishihara Sangyo Kaisha, Ltd.) were used as colorants, and the components shown in Table 3 were used in the composition ratios showed therein. The color tone of composition 8 was adjusted to the same tone as composition 7 by means of using titanium oxide white and titanium oxide yellow. The results are displayed in Table 3.

[0027]

[Table 3]

		Working Example		Comparative Example	
Plate sample		7		8	
Polycarbonate resin (A)	*1	70	parts	70	parts
PBT (B)	*7	30	parts	30	parts
Impact modifier	*8	5	parts	5	parts
Photostabilizer (C)					
	*2	0.04	parts	0.04	parts
	*3	0.04	parts	0.04	parts
Yellow dyestuff (D)					
Disperse Yellow 201	*9	0.13	parts	0.25	parts
ΔYI	200 hrs	-0.7		-3.9	
	400 hrs	-0.5		-6.2	
ΔE^*	200 hrs	2.6		5.3	
	400 hrs	2.3		10.1	

*7: Polybutylene terephthalate (600FP; manufactured by Polyplastics Co., Ltd.)

*8: Impact modifier (S-2001; manufactured by Mitsubishi Rayon Co., Ltd.)

*9: Macrolex Yellow 6G (manufactured by Bayer AG)

[0028] Working Example 4

The same procedure as described in Working Example 1 was employed in full in order to obtain compositions 9 and 10 and plate samples 9 and 10, with the exception that 0.002 parts of Disperse Violet 28 (Sumiplast Violet RR; manufactured by Sumitomo Chemical Co., Ltd.), 0.0006 parts of Solvent Blue 97 (Macrolex Blue RR; manufactured by Bayer AG), and 1.4 parts of titanium oxide white were used as colorants; and the components shown in Table 4 were used in the composition ratios showed therein. The color tone of composition 10 was adjusted to the same tone as composition 9 by means of using Disperse Violet 28, Solvent Blue 97, and titanium oxide white. The results are displayed in Table 4.

[0029]

[Table 4]

		Working Example		Comparative Example	
Plate sample		9		10	
Polycarbonate resin (A)	*1	70	parts	70	parts
PBT (B)	*7	30	parts	30	parts
Photostabilizer (C)					
	*2	0.7	parts	0.7	parts
	*3	0.7	parts	0.7	parts
Yellow dyestuff (D)					
Solvent Green 5	*10	0.0008	parts	0.0003	parts
ΔYI	200 hrs	0.5		4.5	
	400 hrs	1.5		5.2	
ΔE^*	200 hrs	0.9		4.0	
	400 hrs	1.2		5.0	

* 10: Sumiplast Yellow FL7G (manufactured by Sumitomo Chemical Co., Ltd.)

[0030] Working Example 5

The same procedure as described in Working Example 1 was employed in full in order to obtain compositions 11 and 12 and plate samples 11 and 12, with the exception that 0.003 parts of Iron Oxide Red (120ED; manufactured by Toda Kogyo Corporation) and 1.05 parts of titanium oxide white were used as colorants, and the components shown in Table 5 were used in the composition ratios showed therein. The color tone of composition 12 was adjusted to the same tone as composition 11 by means of using Iron Oxide Red and titanium oxide white. The results are displayed in Table 5.

[0031]

[Table 5]

	Working Example		Comparative Example	
Plate sample	11		12	
Polycarbonate resin (A) ^{*1}	70	parts	70	parts
PBT (B) ^{*11}	30	parts	30	parts
Impact modifier ^{*8}	5	parts	5	parts
Photostabilizer (C) ^{*12}	0.5	parts	—	
Yellow dyestuff (D) ^{*13}	0.008	parts	0.008	parts
Pigment Yellow 180				
ΔYI 200 hrs	2.9		4.1	
400 hrs	0.2		6.9	
ΔE^* 200 hrs	2.5		3.9	
400 hrs	0.3		5.5	

*11: Polyethylene terephthalate (NES- 2040; manufactured by Unitika Ltd.)

*12: Benzophenone-based photostabilizer (Sumisorb 130; manufactured by Sumitomo Chemical Co., Ltd.)

*13: PV Fast Yellow HG; manufactured by Hoechst AG

[0032] Working Example 6

The same procedure as described in Working Example 1 was employed in full in order to obtain compositions 13 through 15 and plate samples 13 through 15, with the exception that 0.0062 parts of Disperse Violet 28 (Sumiplast Violet RR; manufactured by Sumitomo Chemical Co., Ltd.), 0.0031 parts of Solvent Blue 97 (Macrolex Blue RR; manufactured by Bayer AG), 1.4 parts of titanium oxide white, and 0.01 parts of carbon black were used as colorants; and the components shown in Table 6 were used in the composition ratios showed therein. The color tones of compositions 14 and 15 were adjusted to the same tone as composition 13 by means of using Disperse Violet 28, Solvent Blue 97, titanium oxide white, and carbon black. The results are displayed in Table 6.

[0033]

[Table 6]

Plate sample	Working Examples		Comparative Example
	13	14	15
Polycarbonate resin (A) *1	70 parts	70 parts	70 parts
PBT (B) *11	30 parts	30 parts	30 parts
Impact modifier *8	5 parts	5 parts	5 parts
Photostabilizer (C) *2	0.5 parts	0.5 parts	0.5 parts
*3	0.5 parts	0.5 parts	0.5 parts
Yellow dyestuff (D) *14	0.003 parts	—	—
Disperse Orange 47 *15	—	0.003 parts	—
Pigment Orange 43			
Yellow pigment			
Solvent Yellow 166 *16	—	—	0.003 parts
ΔYI 200 hrs	2.3	1.3	5.8
400 hrs	1.6	1.0	7.2
ΔE^* 200 hrs	2.0	1.3	4.8
400 hrs	0.5	1.1	5.5

*14: Macrolex Orange R; manufactured by Bayer AG

*15: PV Fast Orange GRL; manufactured by Hoechst AG

*16: Yellow dyestuff that does not whiten when exposed to light (Oraset Yellow GHS; Ciba-Geigy Corporation)

s[0034]

[Effect of the Invention] The polycarbonate resin composition of the present invention exhibits minimal variation in color when exposed to light, retains a stable color tone over prolonged periods, and may be suitably used in outdoor applications or in other environments where exposure to light occurs.

(Continued from front page)

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